

Tailoring the excitation of localized surface plasmon-polariton resonances by focusing radially-polarized beams

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We study the interaction of focused radially-polarized light with metal nanospheres. By expanding the electromagnetic field in terms of multipoles, we gain insight on the excitation of localized surface plasmon-polariton resonances in the nanoparticle. We show that focused radially-polarized beams offer more opportunities than a focused plane wave or a Gaussian beam for tuning the near- and far-field system response. These results find applications in nano-optics, optical tweezers, and optical data storage.

I. INTRODUCTION

Focused radially-polarized beams (FRBs) find application in many fields of optics. For instance, it has been demonstrated that they achieve a tighter focal spot compared to focused plane-wave (FPW) illumination^{1,2}. Moreover, Novotny *et al.* have used them to map the orientation of single molecules³. Van Enk has discussed FRBs in the context of quantum optics for efficient coupling of light to a single emitter⁴. Also in the area of optical tweezers Zhan has shown that metal nanoparticles (NPs) can be trapped in three dimensions under FRBs⁵. Meixner's group has performed a set of experiments to explore the interaction of radially-, azimuthal- and linearly-polarized focused beams with metal NPs^{6,7,8}.

Recently, a number of theoretical works have studied the interaction of tightly-focused beams with spherical NPs. Şendur *et al.* have used diffraction integrals to analyze the near field produced by a silver NP under FPW and FRB illumination⁹. Likewise, van de Nes and Török have developed a generalized Mie theory for Gauss-Laguerre beams, including the case when the NP is displaced from the focus¹⁰. More recently, Moore and Alonso have chosen the multipole expansion approach to develop a generalized Mie theory where the incident beam is a complex focused field. In particular, they study the response of a dielectric NP under FRBs at a fixed wavelength¹¹. Lastly, Lermé *et al.* have studied the extinction of light by silver NPs in the context of the spatial modulation spectroscopy technique¹².

In this paper we extend our earlier study on the localized surface plasmon-polariton (LSP) spectra of metal NPs¹³ to the case of FRBs. Using the multipole expansion approach, we show that, contrary to a FPW¹⁴ and a Gaussian beam¹⁵, FRBs offer more opportunities for manipulating the excitation of higher-order LSP resonances in metal NPs. We discuss these features both in the far- and near-field regimes.

II. GENERALIZED MIE THEORY

Radially-polarized doughnut beams are constructed by superposing the Gauss-Hermite modes with normal po-

larizations¹⁶. The effect of the lens is identified by two parameters, the focusing semi-angle α and the a factor defined as f/w_o ⁴, where f is the focal length of the lens and w_o is the beam waist (see inset in Fig.1). Using the Richards and Wolf formalism for an aplanatic system¹⁷ and adapting it to radially-polarized illumination, the field right after passing the lens is given by $\mathbf{E}(a, \theta) = A \exp(-a^2 \sin^2 \theta) f \sin \theta \sqrt{\cos \theta} \hat{\theta}$, where $\sqrt{\cos \theta}$ represents the apodization function and $\hat{\theta}$ gives the orientation of the electric field in spherical coordinates (r, θ, ϕ) ^{1,18}. Note that this field depends only on a and θ because f can be merged into the amplitude A , which only rescales the intensity of the beam.

The electric field in the image space can now be found by the multipole expansion, where the weight coefficient of each multipole is determined by matching the field at the lens boundary^{13,14,19}. Since the electric field has only a θ component in the far field, the azimuthal number is $m = 0$, and the symmetry around the z axis implies that transverse electric (TE) multipoles have no contribution. The incident field and the coefficients are hence given by

$$\mathbf{E}_{\text{inc}} = \sum_{l=1}^{\infty} B_l \mathbf{N}_{e0l}^{(1)}, \quad (1)$$

$$B_l = \frac{2l+1}{2l(l+1)} \frac{2kf}{i^l \exp(-ikf)} \times \int_0^\alpha |\mathbf{E}(a, \theta)| \frac{dP_l(\cos \theta)}{d\theta} \sin \theta d\theta, \quad (2)$$

where k is the wavevector and $P_l(\cos \theta)$ are Legendre polynomials. Here the multipoles follow the notation of²⁰. The expansion of Eq. (1) is a special case of tightly-focused spirally-polarized beams¹⁹.

Now that the incident field is determined in terms of multipoles, the effect of placing a spherical NP at the origin can be solved by the generalized Mie theory¹³. The scattered and internal fields are respectively found to be

$$\mathbf{E}_s = - \sum_{l=1}^{\infty} a_l B_l \mathbf{N}_{e0l}^{(3)}, \quad \mathbf{E}_i = \sum_{l=1}^{\infty} d_l B_l \mathbf{N}_{e0l}^{(1)}, \quad (3)$$

where a_l and d_l are the Mie coefficients as defined in²⁰. Note that because the incident field does not contain TE

multipoles, these are also absent in the scattered and internal fields. Having the fields expanded in all space, the total scattered (W_s) and extinguished (W_e) powers can also be calculated by integrating the corresponding Poynting vectors in the far field, giving

$$W_s = \frac{\pi}{Zk^2} \sum_{l=1}^{\infty} |B_l|^2 |a_l|^2 \frac{2l(l+1)}{2l+1}, \quad (4)$$

$$W_e = \frac{\pi}{Zk^2} \sum_{l=1}^{\infty} |B_l|^2 \text{Re}\{a_l\} \frac{2l(l+1)}{2l+1}, \quad (5)$$

where Z is the impedance of the background medium. According to Eqs. (4) and (5), W_s and W_e not only depend on the NP size and material, but also vary with the multipole strength (the B_l term)¹³. This clearly shows that for a specific LSP resonance, one can in principle control the scattered and extinguished powers of the corresponding multipole by adjusting the illumination beam and the focusing system.

The cross sections associated with W_s and W_e are found by dividing these quantities by the average intensity incident on the xy section of the NP¹³. This averaging process is necessary because the field in the focal plane is strongly inhomogeneous in contrast to plane-wave illumination. However, the cross sections calculated by this approach lead to unrealistic values that are one or two orders of magnitude larger than those obtained by FPW illumination. The reason behind this delusive behavior is that although the electric field is strong near the focus, the Poynting vector is small and vanishes on axis (see inset in Fig. 2)³. We therefore use another quantity to study the LSP spectra, namely the scattering (\mathcal{K}_s) and the extinction (\mathcal{K}_e) efficiencies, defined by dividing W_s and W_e by the total incident power P_{inc} ²¹: $\mathcal{K}_s = W_s/P_{\text{inc}}$ and $\mathcal{K}_e = W_e/P_{\text{inc}}$. The profiles obtained by this method slightly differ from the cross sections, but still exhibit the resonances of the system.

Many applications of plasmonic materials are based on their near-field properties²², whereas cross sections or efficiencies are far-field quantities. For this reason, we use an alternative definition called the near-field average intensity enhancement K for characterizing the system response. This quantity is simply the total field intensity averaged over the NP surface, divided by the intensity at the origin in the absence of the NP¹³. By taking advantage of the orthogonality of the multipoles, K is separated into radial K_r and tangential K_t components,

$$K_r = \frac{9}{16(kR)^4} \sum_l \left| \frac{B_l}{B_1} \right|^2 \frac{l^2(l+1)^2}{2l+1} \times \quad (6)$$

$$[|\psi_l|^2 + |a_l|^2 |\chi_l|^2 + 2\text{Re}\{a_l^* \chi_l^*\} \psi_l],$$

$$K_t = \frac{9}{16(kR)^2} \sum_l \left| \frac{B_l}{B_1} \right|^2 \frac{l(l+1)}{2l+1} \times \quad (7)$$

$$[|\psi'_l|^2 + |a_l|^2 |\chi'_l|^2 + 2\text{Re}\{a_l^* \chi'_l\} \psi'_l],$$

where R is the NPs radius. In these equations ψ_l and χ_l are respectively the Riccati-Bessel functions of the first

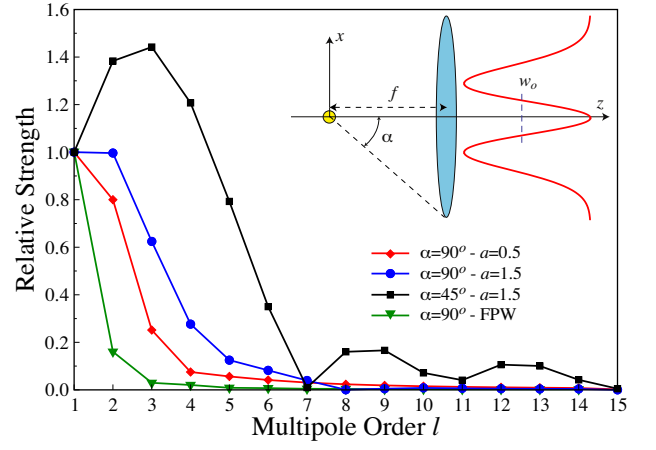


FIG. 1: Relative strength $|B_l/B_1|$ of the multipoles in the incident field. The result for a FPW is also shown for comparison¹³. The inset sketches a radially-polarized beam focused onto a NP. The red curve is the beam intensity profile before the lens, f is the focal length, α the angular semi-aperture, w_o the beam waist and $a = f/w_o$.

and third kinds calculated at the NP surface, and the prime indicates the derivative with respect to kr ^{13,20}.

III. RESULTS AND DISCUSSION

In this section we discuss how FRBs can change the excitation of LSP resonances in metal NPs and its consequences on the far- and near-field optical response. As a case study we focus on the interaction of different types of FRBs with a 100 nm silver spherical NP²³ embedded in glass (background index $n_b = 1.5$). This is a suitable NP size since it is neither too small to eliminate the excitation of higher-order modes, nor too large to suppress the near-field enhancement²⁴. There is a freedom of changing f or w_o to obtain different values of $a = f/w_o$. We set the beam waist to $w_o = 5$ mm and adjust f to select a . We use Mathematica for the calculations²⁵, taking into account up to 15 multipoles for the field expansions.

The relative strength $|B_l/B_1|$ in Fig. 1 shows how much the l^{th} multipole contributes to the fields in comparison to the dipole ($l = 1$). A tightly-focused beam ($\alpha = 90^\circ$ and $a = 0.5$) mainly consists of a dipole and a quadrupole, the latter being 80% of the dipole strength. The next case has again the same focusing angle, but the beam waist is smaller. This is a special illumination because the quadrupole and the dipole coefficients exhibit nearly the same weight. Keeping the same value for a but focusing less tightly ($\alpha = 45^\circ$), the dipole strength becomes even less than the next three multipoles. In summary, the general trend in the incident-wave multipole content is as follows: by increasing the value of α , i.e. focusing more tightly, higher-order modes are suppressed and by increasing the value of a , i.e. longer focal length

or smaller beam waist, higher-order modes get stronger. For the sake of comparison we also plot in Fig. 1 the multipole strength for a FPW. Note that for a FPW and a Gaussian beam the higher-order modes can only be suppressed with respect to an incident plane wave^{13,15}.

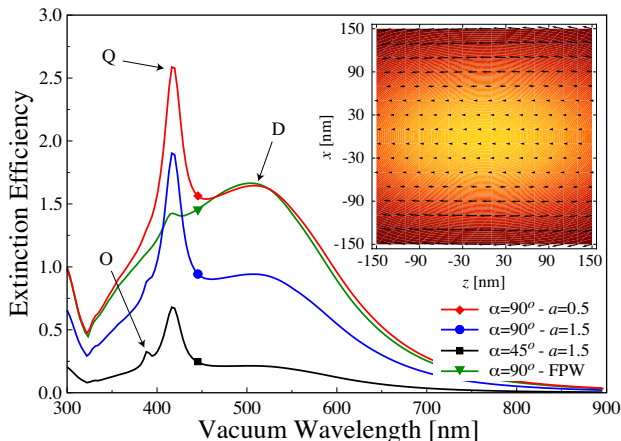


FIG. 2: Extinction efficiency \mathcal{K}_e of a 100 nm silver NP in glass illuminated by three different FRBs and a FPW. D, Q and O respectively label the dipole, quadrupole and octupole resonances. Inset: electric field intensity (contours) and Poynting vector (arrows) in the focal region for $\alpha = 90^\circ$, $a = 1.5$, and $\lambda = 520$ nm.

We next study the far-field LSP spectrum by looking at \mathcal{K}_e (see Fig. 2). The first property that catches one's attention is that \mathcal{K}_e can exceed 2. This does not violate the conservation of energy, because one has to consider the total Poynting vector instead of only the scattered or extinguished components^{20,21}. Indeed the transmitted, reflected, and absorbed powers together are equal to the total input power. The effect of changing the multipoles strength can be observed here by studying the relative heights of the corresponding resonances. For example, the ratios of the quadrupole to dipole peaks are 1.63, 2.04, and 3.22 for the three cases presented in Fig. 1. As expected, the multipole strength determines the relative height of the LSP modes accordingly. Indeed for the FRB with a large content of higher-order multipoles, even the third resonance is clearly excited. To stress the difference with respect to conventional focused light, Fig. 2 shows that almost only the dipole resonance contributes to \mathcal{K}_e for a FPW when $\alpha = 90^\circ$.

Comparing these three cases reveals that the NP extinguishes more power when the beam is focused more tightly. This is simply because more light is concentrated onto the NP. Furthermore, the reason why \mathcal{K}_e is larger for a shorter focal length (smaller a) is that the electric field of the rays farther away from the z axis produce a stronger longitudinal field at the focus¹. The interesting consequence of the multipole expansion is the correlation between the excited LSP resonances and the strength of the corresponding multipoles. This information can be used to control the excitation of each mode in the NP.

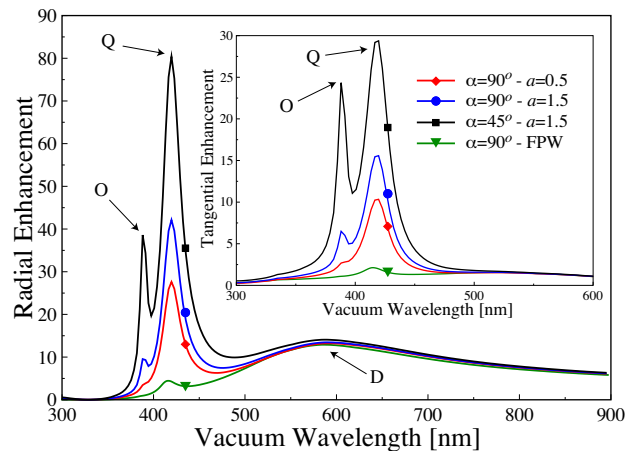


FIG. 3: Radial and tangential (inset) average intensity enhancement for a 100 nm silver NP in glass illuminated by three different FRBs and a FPW. D, Q and O respectively label the dipole, quadrupole and octupole resonances.

We next move our attention to the average near-field intensity enhancement and especially on the radial one (see Fig. 3). For a 100 nm silver NP, the higher-order excited modes yield better K_r and K_t with respect to the dipole one. Moreover, by focusing less tightly or making a larger, they exhibit a stronger enhancement. In comparison to \mathcal{K}_e of Fig. 2, the relative strength of the quadrupole peak with respect to the dipole is larger. This clearly shows the difference between the near- and the far-field regimes^{13,24}. As in the case of FPW illumination, which is shown in Fig. 3 for comparison, the enhancement near the dipole peak remains almost the same irrespective of the focusing parameters. For a FPW K_r and K_t are always less than for plane-wave illumination¹³. On the other hand, with FRBs one can control the enhancement at the quadrupole and higher-order resonances by changing the multipole content of the beam. More precisely, the ratio of quadrupole to dipole peak enhancements is 2.08, 3.13, and 5.71 for the three cases in Fig. 3. For instance, when $\alpha = 45^\circ$ and $a = 1.5$ K_r reaches a factor of 80, which is about 2 and 6 times the value for a plane wave and a FPW for $\alpha = 60^\circ$, respectively¹³.

IV. CONCLUSION

We have studied the near- and far-field response of metal NPs illuminated by FRBs using a 100 nm silver spherical NP in glass as a representative system. The multipole expansion approach allows us to demonstrate the potential of FRBs for tailoring the excitation of LSP resonances and hence controlling the scattering and extinction efficiencies or the near-field enhancement. These findings show that FRBs offer more opportunities than conventional focused light. Our results might find application in the spectroscopy of metal NPs⁸, optical twee-

ers⁵ and optical data storage²⁶. Furthermore, molecular fluorescence can also be improved by FRBs. Since the quadrupole mode exhibits a stronger electric field under appropriate illumination, one could use it to increase the excitation rate and the dipole resonance to amplify the emission rate without the need of designing complicated nanostructures²⁷.

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- ¹ S. Quabis, R. Dorn, M. Eberler, O. Glöckl, and G. Leuchs, "The focus of light – theoretical calculation and experimental tomographic reconstruction," *Appl. Phys. B* **72**, 109–113 (2001).
 - ² R. Dorn, S. Quabis, and G. Leuchs, "Sharper focus for a radially polarized light beam," *Phys. Rev. Lett.* **91**, 233901(4) (2003).
 - ³ L. Novotny, M. R. Beversluis, K. S. Youngworth, and T. G. Brown, "Longitudinal field modes probed by single molecules," *Phys. Rev. Lett.* **86**, 5251–5254 (2001).
 - ⁴ S. J. van Enk, "Atoms, dipole waves, and strongly focused light beams," *Phys. Rev. A* **69**, 043813(8) (2004).
 - ⁵ Q. Zhan, "Trapping metallic Rayleigh particles with radial polarization," *Opt. Express* **12**, 3377–3382 (2004).
 - ⁶ A. V. Failla, H. Qian, H. Qian, A. Hartschuh, and A. J. Meixner, "Orientational imaging of subwavelength Au particles with higher order laser modes," *Nano Lett.* **6** 1374–1378 (2006).
 - ⁷ A. V. Failla, S. Jäger, T. Züchner, M. Steiner, and A. J. Meixner, "Topology measurements of metal nanoparticles with 1 nm accuracy by Confocal Interference Scattering Microscopy," *Opt. Express* **15**, 8532–8542 (2007).
 - ⁸ T. Züchner, A. V. Failla, A. Hartschuh, and A. J. Meixner, "A novel approach to detect and characterize the scattering patterns of single Au nanoparticles using confocal microscopy," *J. Microsc.* **229**, 337–343 (2007).
 - ⁹ K. Şendur, W. Challener, and O. Mryasov, "Interaction of spherical nanoparticles with highly focused beam of light," *Opt. Express* **16**, 2874–2886 (2008).
 - ¹⁰ A. S. van de Nes and P. Török, "Rigorous analysis of spheres in Gauss-Laguerre beams," *Opt. Express* **15**, 13360–13374 (2007).
 - ¹¹ N. J. Moore and M. A. Alonso, "Closed form formula for Mie scattering of nonparaxial analogues of Gaussian beams," *Opt. Express* **16**, 5926–5933 (2008).
 - ¹² J. Lermé, C. Bonnet, M. Broyer, E. Cottancin, S. Marhaba, and M. Pellarin, "Optical response of metal or dielectric nano-objects in strongly convergent light beams," *Phys. Rev. B* **77**, 245406-13 (2008).
 - ¹³ N. M. Mojarad, V. Sandoghdar, and M. Agio, "Plasmon spectra of nanospheres under a tightly focused beam," *J. Opt. Soc. Am. B* **25**, 651–658 (2008).
 - ¹⁴ C. J. R. Sheppard and P. Török, "Efficient calculation of electromagnetic diffraction in optical systems using a multipole expansion," *J. Mod. Opt.* **44**, 803–818 (1997).
 - ¹⁵ J. A. Lock, J. T. Hodges, and G. Gouesbet, "Failure of the optical theorem for Gaussian-beam scattering by a spherical particle," *J. Opt. Soc. Am. A* **12**, 2708–2715 (1995).
 - ¹⁶ R. Oron, S. Blit, N. Davidson, A. A. Friesem, Z. Bomzon, and E. Hasman, "The formation of laser beams with pure azimuthal or radial polarization," *Appl. Phys. Lett.* **77**, 3322–3324 (2000).
 - ¹⁷ B. Richards and E. Wolf, "Electromagnetic diffraction in optical systems II. Structure of the image field in an aplanatic system," *Proc. R. Soc. A* **253**, 358–379 (1959).
 - ¹⁸ K. S. Youngworth and T. G. Brown, "Focusing of high numerical aperture cylindrical-vector beams," *Opt. Express* **7**, 77–87 (2000).
 - ¹⁹ R. Borghi, M. Santarsiero, and M. A. Alonso, "Highly focused spirally polarized beams," *J. Opt. Soc. Am. A* **22**, 1420–1431 (2005).
 - ²⁰ C. F. Bohren and D. R. Huffman, *Absorption and scattering of light by small particles* (Wiley, New York, 1983).
 - ²¹ G. Zumofen, N. M. Mojarad, V. Sandoghdar, and M. Agio, "Perfect reflection of light by an oscillating dipole," *Phys. Rev. Lett.* **101**, 180404(4) (2008) and supplementary material.
 - ²² S. A. Maier and H. A. Atwater, "Plasmonic: Localization and guiding of electromagnetic energy in metal/dielectric structures," *J. Appl. Phys.* **98**, 011101(10) (2005).
 - ²³ P. B. Johnson and R. W. Christy, "Optical constants of the noble metals," *Phys. Rev. B* **6**, 4370–4379 (1972).
 - ²⁴ B. J. Messinger, K. U. von Raben, R. K. Chang, and P. W. Barber, "Local fields at the surface of noble-metal microspheres," *Phys. Rev. B* **24**, 649–657 (1981).
 - ²⁵ Wolfram Research, Inc., *MATHEMATICA*, Version 5.1, Champaign, IL (2004).
 - ²⁶ M. Sugiyama, S. Inasawa, S. Koda, T. Hirose, T. Yonekawa, T. Omatsu, and A. Takami, "Optical recording media using laser-induced size reduction of Au nanoparticles," *Appl. Phys. Lett.* **79**, 1528–1530 (2001).
 - ²⁷ V. Giannini and J. A. Sánchez-Gil, "Excitation and emission enhancement of single molecule fluorescence through multiple surface-plasmon resonances on metal trimer nanoantennas," *Opt. Lett.* **33**, 899–901 (2008).